
Priyadarajan Rây*

Contributions to Chemical Science

Rajarshi Ghosh

Prof. Priyadarajan Rây's contribution to inorganic chemistry and related disciplines is enormous. His work on the elucidation of valency and structure of coordination compounds through magnetic measurements, proposition of Rây–Dutt twist mechanism and the discovery of inner metallic complexes of higher than second-order made him internationally famous. He first reported the two ligand isomers of thiosulphate ion and designed several organic reagents for the detection and estimation of metal ions. Stabilization of unusual oxidation state of metal ions through coordination with suitable ligand was also his important work. Besides science, he was interested in its history. Scientific activities as well as a brief life-sketch of this Indian stalwart of science is summarized in this present endeavour.

1. Introduction

Acharya P. C. Rây wrote in his autobiography [1], "In this connection, the case of Prof. Priyadarajan Rây should have special mention. He is regarded as an acknowledged authority on "complexes and valency" as also on Microchemistry, and it is my practice to submit my own papers to his criticism and judgment before they are contributed to Chemical Societies. My presidential addresses at the annual meetings of the Indian Chemical Society of 1926 and 1929 are based mainly upon his ideas and suggestions. A more silent and unobtrusive worker is seldom to be met with. It was with some difficulty and under pressure that he could be persuaded to go to Europe and give a finishing touch to his attain-



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Keywords

Priyadarajan Rây, coordination complexes, ligand isomerism, thiosulphate, biguanide, inner metallic complex.

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ments. He at any rate, does not seem to suffer from the distemper known as “inferiority complex”. A Rash Behari Ghosh Travelling Fellowship was literally forced on him. He worked for four months in the laboratory of Professor Ephraïmat Berne where he was received and welcomed as a valued colleague whose name and fame had already preceded him. He has already published at least a score of papers, any one of which if submitted to any university will win for him a doctorate. He has not, however, as yet been able to make up his mind to do this.

Events are of two kinds—the silent and the noisy. Priyadarajan’s work come under the former category. Apart from his other contributions, his recent isolation of an isomer of thiosulphuric acid is a singular achievement and marks him out as an original investigator of a very high order.

– Acharya P C Rây

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As evident from the above excerpt, Praiyadarajan Rây was an outstanding inorganic chemist of the early twentieth century India. His international recognition was due to his number of fundamental contributions in inorganic, coordination as well as analytical chemistry. His outstanding scientific efforts under economically and infrastructurally stringent conditions will be regarded forever not only in our country but also abroad.

2. Early Life and Education

Priyadarajan Rây came from a well-known zamindar family in Chittagong (presently in Bangladesh). He was born on 16th January 1888 [2]. His father Kali Kumar Ray was the Sub-deputy Collector of Rangamati (Bangladesh) and mother Sarojini Devi was housewife [2]. After completion of his primary education from the village pathshala, Priyadarajan was admitted to Chittagong Collegiate School. In 1902, when Priyadarajan was a student of class IX, unfortunately, his father passed away. Despite the mental stress, he passed the entrance examination with a first-class and was awarded the ‘Rai Bahadur Golakchandra Scholarship’. Then he joined Chittagong Government College and passed FA therefrom in 1906. Next to this he moved to



Kolkata and joined Presidency College for higher studies. He was a boarder of Eden Hindu Hostel of the College where Dr Rajendra Prasad, the first President of independent India, was his fellow boarder [2]. At that time in Presidency College, he got Acharya J C Bose, Acharya P. C. Ray and many other stalwarts as his teachers. In 1908 and 1911, he completed his BA and MA in Chemistry, respectively. He stood first class first in his MA class and won the University Gold Medal and Motilal Mullik Gold Medal [2].

3. Early Research and Career

Soon after the completion of his post-graduation, Priyadarajan started his research under the supervision of Acharya P. C. Rây. Following year, he published his first research article [3] along with his junior Hemendra Kumar Sen (who later became the Director of Indian Lac Research Institute, Namkum, Ranchi, presently known as Indian Institute of Natural Resins and Gums). In this very year Priyadarajan faced a very serious accident at the Presidency College laboratory. He was adding cuprous thiocyanate to fused potassium thiocyanate in a glass beaker. The process was operated on a strong sulphuric acid bath. Suddenly the beaker cracked exploded, and the heated sulphuric acid fell into Priyadarajan's eyes [2]. He was admitted to Calcutta Medical College immediately where he had to stay for a couple of months for medical treatment. But, despite all the efforts of the physicians, Priyadarajan permanently lost the vision of his left eye [2]. After his release from the hospital, he stayed about two years outside Kolkata to regenerate his lost mental as well as physical strength.

In 1914, he returned to Kolkata and joined City College as the Professor of Chemistry. At that time, Rajabazar Science College of Calcutta University was built under the enthusiastic supervision of the then Vice-chancellor Sir Asutosh Mookerjee. Sir Mookerjee planned to make this college a centre for advanced learning by appointing Professors of Indian origin. Acharya P C Ray joined the Chemistry Department of Rajabazar Science Col-



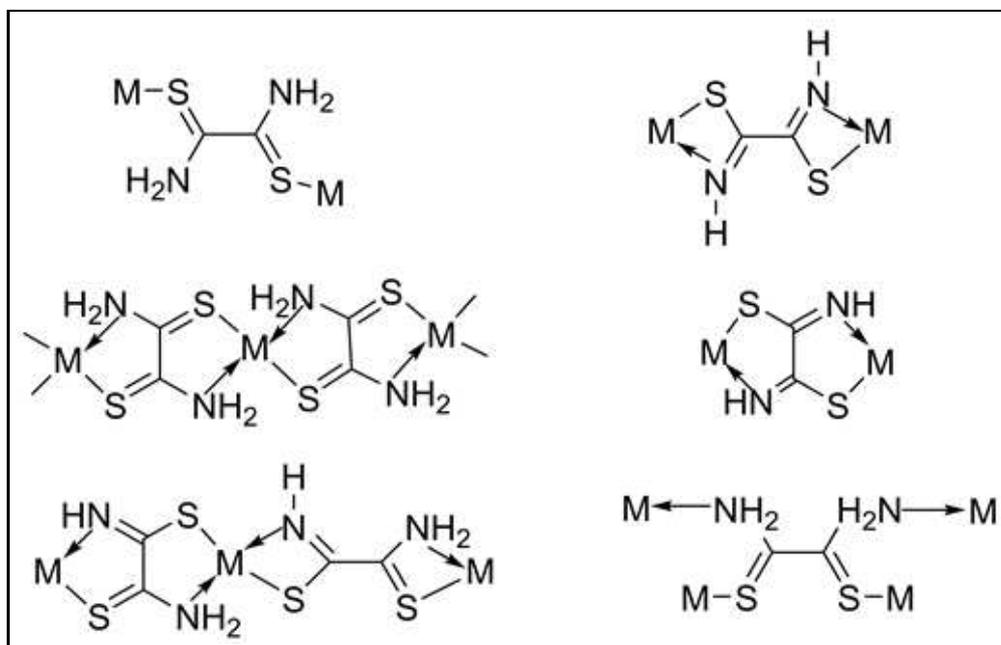
lege as the first Palit Professor after his retirement from the government service at Presidency College. C V Raman joined as the first Palit Professor of Physics there. In 1919, young Priyadarajan joined Calcutta University (Rajabazar Science College) as the Assistant Palit Professor [2]. Joining the University, Priyadarajan immediately focused on executing scientific research. As stated by Acharya Rây [1], Priyadarajan had no affinity for foreign visits, and hence he did not feel the need to have a formal training from abroad to do better in research. It was rightly told that the travelling fellowship of Calcutta University was 'forced' [1] on him. In 1929, he went to Switzerland to work with Professor Fritz Ephraim [2]. Next year, he visited the laboratory of the famous Austrian microchemist Professor Frederic Emich [2]. After visiting different laboratories in several other countries like Germany, France, Czechoslovakia, Hungary, Holand and England, he returned to India in 1930 [2]. In 1937 he was promoted as Khaira Professor of Chemistry in Calcutta University and in 1946, after the sad demise of Acharya Rây, he became Palit Professor [2].

4. Contributions to Chemical Sciences

Prof. Priyadarajan Rây made many significant contributions to inorganic chemistry viz, design and synthesis of new ligands and their coordination compounds, determination of stability of different unusual oxidation levels of metals, the structure of the coordination complexes, their stability, optical activity, reaction mechanism, etc.

Prof. Priyadarajan Rây made many significant contributions to inorganic chemistry viz, design and synthesis of new ligands and their coordination compounds, determination of stability of different unusual oxidation levels of metals, the structure of the coordination complexes, their stability, optical activity, reaction mechanism, etc. He also designed several methods for the detection and estimation of different metal ions in macro and micro amounts. Before going to the details of his work, it is noteworthy to mention that Priyadarajan had a great nationalistic feeling. After the establishment of the Indian Chemical Society in 1924 under the presidentship of Acharya P C Rây, most of his publications appeared in the journal of the society (*Journal of Indian Chemical Society*) [4].





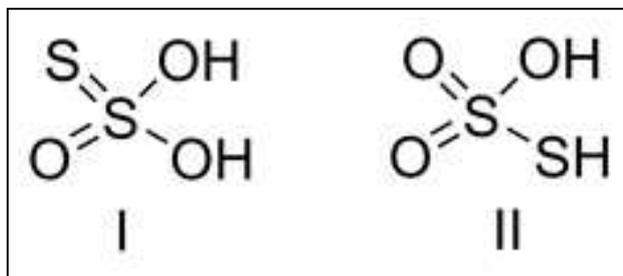
4.1 Design of Organic Reagents in Inorganic Chemistry

Figure 1. Few coordination modes of rubeanic acid.

For the detection and estimation of metal ions using volumetric, gravimetric and spectrophotometric methods, Prof. Rây designed several organic reagents like rubeanic acid and its derivatives, quinaldinic acid, dimercaptothiobiazone (or bismuthiol-I), biguanide, cystin and many others (like amidoximes, hydroxamic acid, etc). In a faintly ammoniacal medium, rubeanic acid, as observed by Prof. Rây, can detect Cu(II) even at the concentration of 30 ppm (1 ppm = 1 mg/L or 10^{-6} g/mL) [5]. The resulting structure of the mixture of Cu(II) and rubeanic acid was discovered later [6]. Rubeanic acid having its different coordination modes (Figure 1) forms a chain-like structure with Cu(II) [6]. The idea of *in situ* generation of ammonia (using urotropin) as precipitating agents by Prof. Rây [7] was quite new then.

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Figure 2. Ligand isomers of $\text{H}_2\text{S}_2\text{O}_3$.



4.2 Ligand Isomerism of Thiosulphate

Priyadarajan Rây isolated two different isomeric modifications of a compound tetrahydrogen pentacyano-thiosulphato-cobalt(III) $\text{H}_4[\text{Co}(\text{CN})_5(\text{S}_2\text{O}_3)]$ [8]. These on hydrolysis, produce different products. One isomer generates sulphur dioxide and elemental sulphur as usual, and the other generates hydrogen sulphide and sulphuric acid. This observation led him to conclude that the same ligand (thiosulphate) in two complexes are in isomeric forms [8]. The respective acids of the ligand (thiosulphuric acid) will be of the forms as shown in *Figure 2*.

4.3 Magnetochemistry

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Systematic investigation of valence state (oxidation level) of the central metal ion, nature of chemical bond and stereochemistry of coordination complexes using magnetic susceptibility data was very intelligently investigated by Prof. Priyadarajan Rây. His expertise in this then-new field of chemical research made him famous around the globe. In 1968, Priyadarajan was invited to deliver the Shanti Swarup Bhatnagar memorial lecture at the National Institute of Sciences in India (presently Indian National Science Academy). The more than twenty pages Bhatnagar memorial lecture [9] by Prof. Rây remains a classic even today in the field of magnetic measurements for the elucidation of the molecular structure of transition metal complexes.

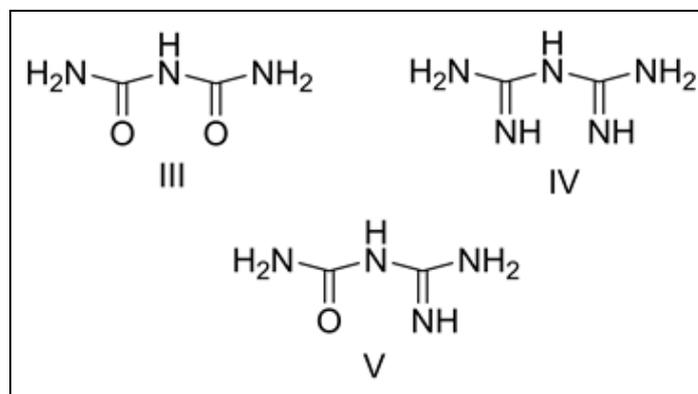
Debendra Mohan Bose, the then Ghosh Professor of Physics, Calcutta University (the second Director (next to Acharya J C Bose)

of Bose Institute, Kolkata) was interested in magnetism. On request of Prof. Bose, Priyadarajan prepared several coordination complexes whose magnetic behaviour were studied by the former. Though they did not have any joint publication, in his famous *Philosophical Magazine* paper in 1928, Prof. Bose clearly stated [10], "All these compounds, with the exception of the last two, have been prepared in the Science College by Messrs. P Ray and H G Bhar, with whose permission their results are given here." From these data of magnetochemical aspects, Priyadarajan perhaps published his first scientific paper [11] on magnetochemistry. In those days, Prof. S S Bhatnagar from the University of Punjab, Lahore (undivided India), made similar important contributions to the magnetostructural correlation of metallic complexes [12]. His contributions to the construction of a sensitive Gouy magnetic balance deserves special mention [13].

Linus Pauling, on the basis of quantum mechanical considerations, published his valence bond theory [14] which proposed the idea of hybrid bonding orbitals to account the magnetic properties and structure of transition metal complexes. Using the idea of inner orbital and outer orbital complexes, Priyadarajan investigated a large number of complex compounds to explain their valency, bonding and structure. Here, it is to note that the terms 'inner orbital' and 'outer orbital' complexes were coined by Henry Taube [15] in 1952. 'Penetration' and 'associated' complexes were called to the similar type of compounds, respectively by Priyadarajan Rây much earlier [9]. In 1941, Prof. Rây delivered the Coochbehar Professorship lecture (constituted by the generous donation of Maharaja of Coochbehar, India) at the Indian Association for the Cultivation of Science (IACS), Kolkata. Out of these deliberations, the book *The Theory of Valency and the Structure of Chemical Compounds* (published by IACS) was composed. The book became extremely popular throughout the globe and was reviewed in *Nature* [16]. After going through these lectures, the Nobel laureate Prof. Linus Pauling became interested in the research of Priyadarajan Rây, and during his visit to India in 1955, Prof. Pauling met with Priyadarajan in Kolkata

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Figure 3. Biurate (III), Biguanide (IV) and Dicyandiamidine (V).



and had a wonderful exchange of scientific thoughts.

4.4 Chemistry of Biguanide Ligand

Biurates (III, *Figure 3*) have long been known as a good chelating agent. Biguanide (BigH) and dicyandiamidine are different derivatives of biurates (IV and V, *Figure 3*) which were also found to have excellent chelation properties. Prof. Priyadarajan Rây elaborately investigated the synthesis, characterization, and applications of metal complexes of a biguanide and related ligand systems. His whole work was summarized by himself in a review [17] which was highly cited later. Metallic complexes of biguanides were first reported by F Emich (with whom Priyadarajan Rây worked abroad) and A Friedrich even before the discovery of coordination complexes by Alfred Werner (Ref. Nos. 49–51 and 53 in [15]).

Several BigH complexes of Cu(II), Ni(II), Pd(II), Ag(III), Mn(III), Mn(IV), V(IV), Re(V), Os(VI), Cr(III), and Zn(II) were reported by Prof. Rây.

BigH chemistry was largely extended by Prof. Priyadarajan Rây in different fundamental aspects as mentioned below.

4.4.1. Inner metallic complexes of third and fourth order: The metal complexes where the charge of the metal ion and its coordination number are satisfied either partially or fully are called inner

Prof. Priyadarajan Rây elaborately investigated the synthesis, characterization, and applications of metal complexes of a biguanide and related ligand systems.

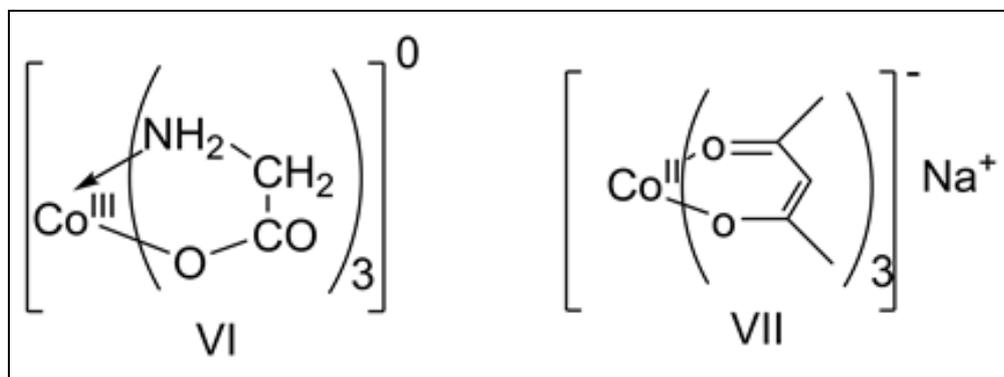


Figure 4. Inner metallic complexes of first and second order.

metallic complexes or inner complexes. When the ligand compensates both the charge and coordination number of the metal ion, the complexes are called inner metallic complexes of the first order. Example is $[\text{Co}^{\text{III}}(\text{gly})_3]^0$ (gly = glycine) (VI, Figure 4). Again, when either the coordination number or the charge is not satisfied fully in the metal complexes, these are called inner metallic complexes of the second order. These types of complexes are often ionic in nature, e.g. $\text{Na}[\text{Co}^{\text{II}}(\text{acac})_3]$ (acacH = acetylacetone) (VII, Figure 4).

Inner metallic complexes of the third and fourth-order were discovered by Prof. Priyadarajan Rây [18]. BigH (IV, Figure 3) is a monobasic bidentate ligand. The two imine nitrogens ($=\text{NH}$) of BigH coordinate to the metal centre to form a metal complex. If the proton of the secondary amine nitrogen in BigH is deprotonated using a suitable anhydrobase, the ligand becomes monoanionic. The *bis* complexes of bivalent metal and this monoanionic biguanide (Big) (VIII, Figure 5) are electrically neutral and insoluble in polar solvents. When the uni negative N donor of BigH in this particular complex is carefully protonated, the complex becomes ionic (IX, Figure 5). This type of complex (VIII, Figure 5) with anionic ligand (which can be protonated on its complexed form) was called an inner metallic complex of the third-order by Priyadarajan Rây.

Again, if the ligand in an inner metallic complex of the first-order (X, Figure 5) contains both Lewis acidic and basic groups, then

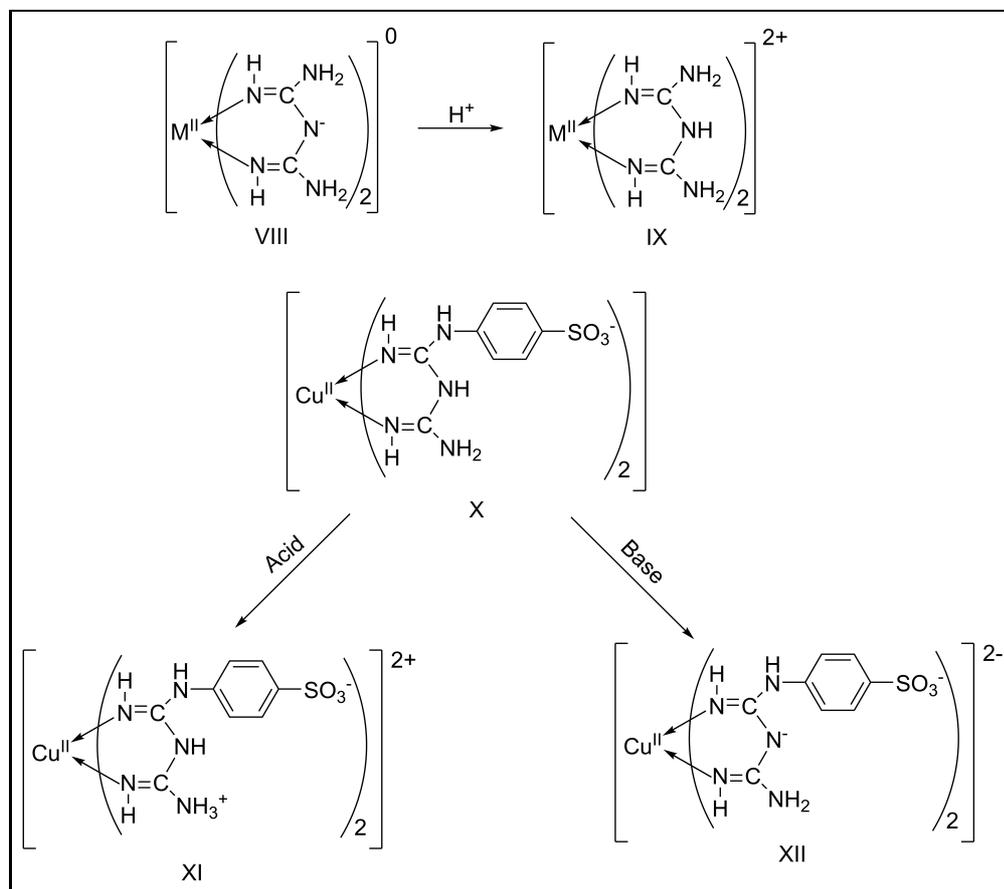
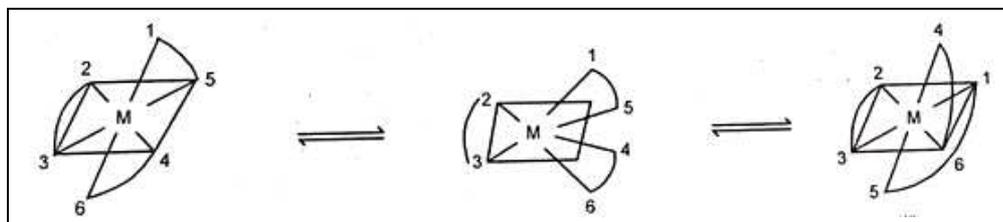


Figure 5. Inner metallic complexes of third and fourth order.

on the treatment of acid, the basic part(s) gets (get) protonated and the complex becomes cationic (inner metallic complexes of second-order, XI, Figure 5). Similarly, on the treatment of base, the proton(s) from the acidic part(s) gets (get) deprotonated making the complex anionic (inner metallic complexes of second-order, XII, Figure 5). Prof. Priyadarajan Rây termed the starting ampholytic complex type (X, Figure 5) as the inner metallic complex of fourth-order.

4.4.2 Rây–Dutt twist mechanism: A twist mechanism (rhombic or tetragonal) [19] was first put forward by Profs. Priyadarajan Rây and Nihar Kumar Dutt [20] (then a young faculty member at Rajabazar Science College, Calcutta University) to explain the



racemization of tris(BigH)cobalt(III) ion, $[\text{Co}(\text{BigH})_3]^{3+}$. This complex has sufficient thermodynamic and kinetic stability. Hence the racemization is operated, according to Rây and Dutt, through the distortion of the structure rather than one-ended dissociation of the chelate ring (Figure 6). Very soon, the work became popular at international level and the mechanism was named as 'Rây-Dutt twist'. Nowadays, this mechanism is incorporated in the undergraduate chemistry syllabus of almost all the universities. Later, another type of twist mechanism (trigonal) was proposed by J C Bailar in 1958 [21]. This mechanism is now known as 'Bailar twist'.

Figure 6. Rây-Dutt twist mechanism.

In the early 1970s, Prof J C Bailar visited IACS [22]. During his visit, he went to the Hindustan Park residence (in Kolkata) of Prof. Priyadarajan Rây to meet him. Prof. Rây's health was not good enough then, and he was in a half-sitting posture on his bed when Prof. Bailar arrived. Entering his room, astonishingly, Prof. Bailar did not receive a chair, instead he sat on a mat on the floor with his folded legs [22] and started chatting with Priyadarajan modestly. Unhesitatingly, the regard and courtesy conferred by Prof. Bailar to Priyadarajan on that very day should be an instance in Indian science.

Priyadarajan Rây did a lot of work on the higher oxidation state of metals and some other aspects of inorganic as well as coordination chemistry. His work on stabilization of Ni(IV) [25] was very popular. He stabilized nickel as alkali nickel periodate MNiO_6 (M = alkali metal ion).

4.5 Some Other Work

Besides these, Priyadarajan Rây did a lot of work on the higher oxidation state of metals and some other aspects of inorganic as well as coordination chemistry. His work on stabilization of Ni(IV) [23] was very popular. He stabilized nickel as alkali nickel periodate MNiO_6 (M = alkali metal ion). As mentioned pre-

viously, Priyadarajan termed this very complex as a 'penetration complex' which was later known as the inner orbital complex. Moreover, he stabilized Ag(II) and Ag(III) with suitable ligands. In case of high valent metal chemistry, a few reports before Priyadarajan Rây was found [24].

His work on the polyhalogen compounds of hydrogen [25] was also important. In 1940, Priyadarajan showed in collaboration with Prof. S D Chatterjee (then a student of Prof. D M. Bose at Bose Institute, and later Professor of Physics, Calcutta and Jadavpur Universities [26]) that Werner's primary and secondary valencies are indistinguishable in HgI_4^{2-} and BiI_4^{2-} [27]. He showed this using radioactive iodine. In microchemistry, Prof. Priyadarajan Rây was an authority in our country. He designed several experiments for the detection and estimation of metal ions using a micro amount of chemical reagents.

4.6 Work on History of Science and Popular Science Writing

Along with his research in fundamental science, Prof. Priyadarajan Rây was interested in its history; particularly the history of science in ancient India. He had written several articles in Bengali and English on this very topic.

Along with his research in fundamental science, Prof. Priyadarajan Rây was interested in its history; particularly the history of science in ancient India. He had written several articles in Bengali and English on this very topic. Among those, his notable work was the publication of the revised edition of *History of Hindu Chemistry* by Acharya P C Rây under the title *History of Chemistry in Ancient and Medieval India* [28]. Besides, he had written several popular science articles in different Bengali journals/periodicals like *Prabasi*, *Jnan-O-Bijnan* (published by Bangiya Bijnan Parishad), *Desh*, etc.

5. Conclusion

In his personal life, Prof. Priyadarajan Rây was honest and led a very simple life. He also had an acute yearning of spirituality. He had regular correspondence with some of the spiritual leaders like Srimat Anirban and Dilip Kumar Roy (son of famous Bengali poet and dramatist Dwijendralal Roy). An important fact that



highlights Prof. Priyadarajan Rây's principles must be cited. He was appointed as the Director (officiating) of IACS in 1956, after the sudden demise of the then Director Prof. Megh Nad Saha. As long as Prof. Rây held the post, he received only Re. 1/- per month as the remuneration [22]. Building the boys' research scholar hostel of IACS, a long-standing demand of the students, was made possible utilizing the unreceived remuneration grant of Prof. Rây [22].

His Himalayan contributions to the fields of inorganic, coordination as well as analytical chemistry, have been crucial in the gradual development of the respective subjects. He was honoured with several fellowships and memberships of various scientific societies of international repute. To name a few, he was the Foundation Fellow (elected in 1935) of the National Institute of Sciences in India (presently Indian National Science Academy) and the Indian Chemical Society (elected in 1924). He was elected as the President of the Society during 1947–48. He was the President of the Chemistry section of Indian Science Congress held in Bengaluru in 1932. The Universities of Calcutta, Jadavpur, and Burdwan bestowed him with doctorate of science (*honoris causa*). Moreover, he was invited to talk at different foreign institutions at different times. However, he never left the country as he was deadly against the publicity of his own work. Acharya P C Rây had aptly called him (Priyadarajan) a “silent” [1] worker. Prof. Rây passed away on 11 December 1982. While it is over thirty years since his demise, his dedication to research, love for his motherland, and contributions to science remains exemplary and will be revered forever. Students and teachers, generations after generations, will remember him through his voluminous work.

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